



AFRL-RX-WP-JA-2018-0045

**TEMPERATURE DEPENDENT SELLMEIER
EQUATION FOR THE REFRACTIVE INDEX OF GAP
(POSTPRINT)**

**Jean Wei, Joel M. Murray, and Jacob O. Barnes
UES, Inc.**

**Shekhar Guha
AFRL/RX**

**Douglas M. Krein
General Dynamics Institute of Technology**

**Peter G. Schunemann
BAE Systems**

**30 JANUARY 2018
Interim Report**

**Distribution Statement A.
Approved for public release: distribution unlimited.**

© 2018 OPTICAL SOCIETY OF AMERICA

**(STINFO COPY)
AIR FORCE RESEARCH LABORATORY
MATERIALS AND MANUFACTURING DIRECTORATE
WRIGHT-PATTERSON AIR FORCE BASE, OH 45433-7750
AIR FORCE MATERIEL COMMAND
UNITED STATES AIR FORCE**

REPORT DOCUMENTATION PAGE				Form Approved OMB No. 0704-0188	
<p>The public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Department of Defense, Washington Headquarters Services, Directorate for Information Operations and Reports (0704-0188), 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number. PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ADDRESS.</p>					
1. REPORT DATE (DD-MM-YY) 30 January 2018		2. REPORT TYPE Interim		3. DATES COVERED (From - To) 30 July 2015 – 30 December 2017	
4. TITLE AND SUBTITLE TEMPERATURE DEPENDENT SELLMEIER EQUATION FOR THE REFRACTIVE INDEX OF GAP (POSTPRINT)				5a. CONTRACT NUMBER FA8650-15-C-5071	
				5b. GRANT NUMBER	
				5c. PROGRAM ELEMENT NUMBER 65502F	
6. AUTHOR(S) 1) Jean Wei, Joel M. Murray and Jacob O. Barnes – UES, Inc. 2) Shekhar Guha – AFRL/RX (continued on page 2)				5d. PROJECT NUMBER 3005	
				5e. TASK NUMBER	
				5f. WORK UNIT NUMBER X10W	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) 1) UES, Inc. 4401 Dayton-Xenia Rd. Dayton, OH 45432 2) AFRL/RX Wright-Patterson AFB Dayton, OH 45433 (continued on page 2)				8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Air Force Research Laboratory Materials and Manufacturing Directorate Wright-Patterson Air Force Base, OH 45433-7750 Air Force Materiel Command United States Air Force				10. SPONSORING/MONITORING AGENCY ACRONYM(S) AFRL/RXAP	
				11. SPONSORING/MONITORING AGENCY REPORT NUMBER(S) AFRL-RX-WP-JA-2018-0045	
12. DISTRIBUTION/AVAILABILITY STATEMENT Distribution Statement A. Approved for public release; distribution unlimited.					
13. SUPPLEMENTARY NOTES PA Case Number: 88ABW-2018-0054; Clearance Date: 31 Dec 2017. This document contains color. Journal article published in Optical Materials Express, Vol. 8, No. 2, 1 Feb 2018. © 2018 Optical Society of America. The U.S. Government is joint author of the work and has the right to use, modify, reproduce, release, perform, display, or disclose the work. The final publication is available at www.osapublishing.org https://doi.org/10.1364/OME.8.000485					
14. ABSTRACT (Maximum 200 words) A temperature-dependent Sellmeier equation for GaP, valid for wavelengths between 0.7 and 12.5 μm over a temperature range of 78 to 450 K, is presented. The temperature dependence values of the generated wavelengths in nonlinear frequency conversion calculated using this equation match well the experimentally measured values.					
15. SUBJECT TERMS Nonlinear optical materials; Harmonic generation and mixing; CdSiP ₂ ; Sellmeier equation; crystal temperature					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT: SAR	18. NUMBER OF PAGES 9	19a. NAME OF RESPONSIBLE PERSON (Monitor) Thomas Cooper 19b. TELEPHONE NUMBER (Include Area Code) (937) 255-9620
a. REPORT Unclassified	b. ABSTRACT Unclassified	c. THIS PAGE Unclassified			

REPORT DOCUMENTATION PAGE Cont'd

6. AUTHOR(S)

- 3) Douglas M. Krein - GDIT
- 4) Peter G. Schunemann – BAE, Inc.

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

- 3) General Dynamics Information Technology
2673 Commons Blvd Ste 200
Beavercreek, OH 45431
- 4) BAE Systems
144 Daniel Webster Hwy 24
Merrimack, NH 03054



Temperature dependent Sellmeier equation for the refractive index of GaP

JEAN WEI,^{1,2} JOEL M. MURRAY,^{1,2} JACOB O. BARNES,^{1,2} DOUGLAS M. KREIN,^{2,3} PETER G. SCHUNEMANN,⁴ AND SHEKHAR GUHA^{2,*}

¹UES, Inc., Dayton, Ohio 45432, USA

²Air Force Research Laboratory, Wright Patterson Air Force Base, Ohio 45433, USA

³General Dynamics Information Technology, Dayton, Ohio 45431, USA

⁴BAE Systems, Nashua, New Hampshire 03054, USA

*shekhar.guha@us.af.mil

Abstract: A temperature-dependent Sellmeier equation for GaP, valid for wavelengths between 0.7 and 12.5 μm over a temperature range of 78 to 450 K, is presented. The temperature dependence values of the generated wavelengths in nonlinear frequency conversion calculated using this equation match well the experimentally measured values.

© 2018 Optical Society of America under the terms of the [OSA Open Access Publishing Agreement](#)

OCIS codes: (160.4330) Nonlinear optical materials; (190.2620) Harmonic generation and mixing; (140.3070) Infrared and far-infrared lasers.

References and links

1. P. J. Dean, G. Kaminsky, and R. B. Zetterstrom, "Intrinsic Optical Absorption of Gallium Phosphide Between 2.33 and 3.12 eV," *J. Appl. Phys.* **38**(9), 3551–3556 (1967).
2. D. F. Parsons and P. D. Coleman, "Far Infrared Optical Constants of Gallium Phosphide," *Appl. Opt.* **10**(7), 1683 (1971).
3. P. G. Schunemann, L. A. Pomeranz, D. J. Magarrell, J. C. McCarthy, K. T. Zawilski, and D. E. Zelmon, "1064-nm-Pumped Mid-Infrared Optical Parametric Oscillator Based on Orientation-Patterned Gallium Phosphide (OP-GaP)," in *CLEO: 2015*, OSA Technical Digest (online) (Optical Society of America, 2015), paper SW3O.4.
4. D. J. Creeden, P. Schunemann, L. Pomeranz, K. Snell, and S. D. Setzler, "Long-wave Infrared Parametric Generation and Amplification in Orientation Patterned GaP" in *Advanced Solid State Lasers*, OSA Technical Digest (2016), paper ATu5A.
5. P. G. Schunemann, L. A. Pomeranz, and D. J. Magarrell, "First OPO Based on Orientation-Patterned Gallium Phosphide (OP-GaP)," in *CLEO: 2015*, OSA Technical Digest (Optical Society of America, 2015), paper SW3O.1.
6. A. N. Pikhtin, V. T. Prokopenko, and A. D. Yas'kov, "Dispersion of the refractive index of light and permittivity of gallium phosphide," *Sov. Phys. Semicond.* **10**, 1224–1226 (1976).
7. D. E. Zelmon et al., Ref. 7 of [3].
8. T. S. Moss, S. D. Smith, and T. D. F. Hawkins, "Absorption and Dispersion of Indium Antimonide," *Proc. Phys. Soc. B* **70**(8), 776–784 (1957).
9. T. Skauli, P. S. Kuo, K. L. Vodopyanov, T. J. Pinguet, O. Levi, L. A. Eyres, J. S. Harris, M. M. Fejer, B. Gerard, L. Becouarn, and E. Lallier, "Improved dispersion relations for GaAs and applications to nonlinear optics," *J. Appl. Phys.* **94**(10), 6447–6455 (2003).
10. G. A. Slack and S. F. Bartram, "Thermal expansion of some diamondlike crystals," *J. Appl. Phys.* **46**(1), 89–98 (1975).
11. K. Haruna, H. Maeta, K. Ohashi, and T. Koike, "The negative thermal expansion coefficient of GaP crystal at low temperatures," *J. Phys. Chem.* **19**, 5149 (1986).
12. K. Kato, N. Umemura, and V. Petrov, "Sellmeier and thermo-optic dispersion formulas for CdSiP₂," *J. Appl. Phys.* **109**(11), 116104 (2011).
13. M. Born and E. Wolf, *Principles of Optics*, 7th ed. (Cambridge University Press, 1999).

1. Introduction

Orientation patterned GaP (OP-GaP) has been of increasing recent interest for quasi-phase-matched (QPM) nonlinear optical frequency mixing due to several attractive properties of GaP, including high thermal conductivity, wide transparency extending into the visible wavelength range and low linear and nonlinear absorption at various wavelengths of interest due to its large direct bandgap energy (2.87 eV) [1]. For accurate determination of the

requisite grating spacing for QPM interactions, the refractive index values at the interacting wavelengths need to be known with high accuracy. Since different wavelengths can be potentially generated from the same crystal by changing the temperature of the crystal, it is also important to know the refractive index values at different temperatures.

Measurements of the refractive index of GaP at room temperature over a wide wavelength range spanning the visible to microwave wavelengths has been reported [2]. From the frequency dependent expression for the dielectric constant given in Ref. [2], a Sellmeier equation with four poles has been derived for GaP:

$$n^2(\lambda) = 1 + \frac{A_1\lambda^2}{\lambda^2 - A_2} + \frac{B_1\lambda^2}{\lambda^2 - B_2} + \frac{C_1\lambda^2}{\lambda^2 - C_2} + \frac{D_1\lambda^2}{\lambda^2 - D_2} \quad (1)$$

where the wavelength λ is in microns, and $A_1 = 1.39$, $A_2 = 0.172$, $B_1 = 4.131$, $B_2 = 0.234$, $C_1 = 2.57$, $C_2 = 0.345$, $D_1 = 2.056$, and $D_2 = 27.5$. Three of the poles are in the ultraviolet region and the pole D_2 is in the long wave infrared.

Experimental results are available in the literature describing the frequency conversion of pump wavelengths of 1.064 μm , 1.559 μm , and 2.09 μm in orientation patterned GaP (OP-GaP) crystals fabricated with grating spacings of 20.8 μm , 61.1 μm , and 92.7 μm , respectively, based on the index values given in Eq. (1) [3–5]. The signal and idler wavelength pairs (in μm) predicted by Eq. (1) for these grating spacings and pump wavelengths are (1.388, 4.752), (1.928, 8.137), and (3.550, 5.076), respectively, and their experimentally measured values are respectively (1.385, 4.591), (1.923, 8.236), and (3.54, 5.1). The predicted and experimentally observed values are close, indicating that the room temperature index values obtained from Eq. (1) are reliable as a starting point.

The dispersion formula for refractive index of GaP was also given in Ref. [6]. (at 297 K and 105 K) in the ‘Pikhtin form’. The grating spacings for quasi-phase matched three-wave mixing predicted by this formula were far from the experimentally measured values. By contrast, the grating spacings Λ predicted by Eq. (1) were different from the experimental measurements by less than 1%. (See Table 1.)

Table 1. Experimental wavelengths and grating spacings, and predicted grating spacings

Pump (μm)	Signal (μm)	Idler (μm)	Λ_{Pikhtin} (μm)	$\Lambda_{\text{Eq. (1)}}$ (μm)	$\Lambda_{\text{Exp.}}$ (μm)
1.064	1.385	4.591	14.3	20.6	20.8
1.559	1.923	3.54	63.7	61.3	61.1
2.09	8.236	5.1	120.5	92.8	92.7

A temperature dependent Sellmeier equation for GaP was previously obtained from refractive index values measured with a prism [7]. However these measurements were performed over a limited temperature and wavelength range (295 to 395 K, 1 to 5.5 μm).

2. Measurement technique

To obtain the refractive indices of GaP over a wide range of temperature and wavelength, the method outlined by Moss [8] and used more recently for accurate measurement of refractive indices of GaAs [9] was used. A single crystal, (100)-oriented wafer of GaP grown by the Bridgman technique was polished on both sides and thinned to 158 ± 2 μm . Transmission spectra of the samples were taken on a Perkin Elmer FTIR spectrometer run with a step size of 0.125 cm^{-1} over the wavenumber range of 16,667 to 400 cm^{-1} (i.e., wavelength range of 0.67 to 25 μm) and over a temperature range of 78 K to 450 K.

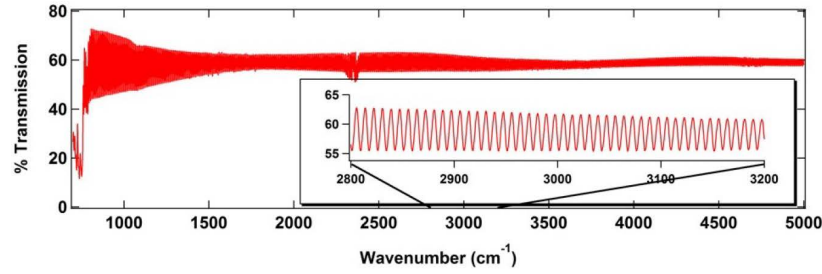


Fig. 1. Transmission spectrum of a GaP wafer at 295 K.

A representative spectrum taken at room temperature is shown in Fig. 1, over a limited spectral range for clarity. The spectra showed a series of fringes corresponding to constructive and destructive interference. If d denotes the sample thickness, then constructive interference at a wavelength λ_m indicates the relationship:

$$2n(\lambda)d = m\lambda \quad (2)$$

where the fringe number m is an integer and $n(\lambda_m)$ is the value of the refractive index at λ_m . Assuming the 295 K value of the refractive index to be known from literature [2] at a long wavelength, say around $10\text{ }\mu\text{m}$ where the fringes are relatively sparse, the fringe number m at that wavelength can be accurately determined from Eq. (1), even with the uncertainty of $2\text{ }\mu\text{m}$ in the wafer thickness. From the value of m at a given fringe wavelength, the fringe numbers at all the shorter and longer wavelengths at which the fringes appeared are obtained. For discrete values of d within its measured limits ($\pm 2\text{ }\mu\text{m}$), the values of $n(\lambda_m)$ are obtained from Eq. (2) at the values of the fringe maxima, and then at arbitrary wavelengths through spline interpolation.

Several GaP samples were studied. Ideally, if the samples were identical in all but thickness, the $n(\lambda)d$ product determined from the measured fringe spectra (using Eq. (2), with known m) for any one sample would match the same product (to within a multiplicative constant) for any other sample of the same material, across the spectrum. This was not found to be the case, indicating that the refractive index is very sensitive to the concentration of sample impurities and/or crystal defects. The correct refractive index values for intrinsic GaP at room temperature were taken to be those given by Eq. (1), and these were compared to the $n(\lambda)d$ product from the GaP sample having the best spectral fringe contrast. If the refractive index of the GaP sample reported here matched that of the samples studied by Parsons and Coleman [2] and Bond [cited in Ref. [2]], then the ratio $(n(\lambda)d)_{\text{FTIR}} / n(\lambda)_{\text{Eq. (1)}}$ would simply equal a constant d , representing the sample thickness. As it was, the dispersion curve deviated somewhat from Parsons and Coleman's result, so that the ratio was equal to a slightly wavelength-dependent function $d(\lambda)$ (see Fig. 2). The variation however was not large: its maximum extent over the wavelength range of 0.7 to $12\text{ }\mu\text{m}$ was $\sim 200\text{ nm}$, or $\sim 0.1\%$.

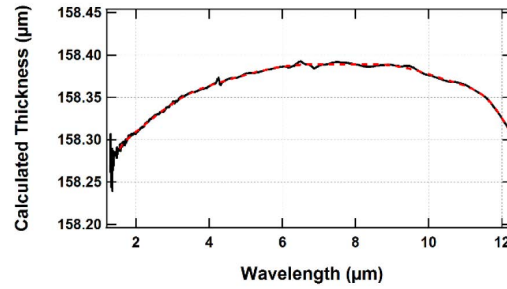


Fig. 2. Calculated sample thickness at 295 K, dividing $n(\lambda)d$ product (from spectral data) by refractive index values from Eq. (1).

From the values of $d(\lambda)$, thicknesses at other temperatures were then calculated using the known temperature-dependent thermal expansion coefficient [10, 11]. Spectra at these other temperatures yield fringe orders m and the wavelengths λ_m corresponding to the transmittance maxima, and from these and Eq. (2), the refractive indices at other temperatures are obtained, referenced to the samples and measurements of Parsons and Coleman and Bond at 300 K. (The nominal “300 K” mentioned in Ref. [2] was assumed here to be “room temperature,” and the spectral data taken here at 295 K was matched to Eq. (1).)

3. Results and discussion

From the FTIR spectra of the GaP sample, the refractive index values were obtained over a temperature range of 78 K to 450 K and wavelength range of 0.7 to 12.5 μm , and the dispersion curves are shown in Fig. 3(a). Fitting the values of n to Eq. (1), the Sellmeier coefficients (A_1, \dots, D_2) were found for each measured temperature. However, the temperature dependence of these coefficients could not be fit using simple polynomial expressions. Using instead an equivalent form of the Sellmeier equation (as used in Ref. [12] and discussed in Ref. [13] on p. 99, Eq. (38)):

$$n^2 = A(T) + \frac{B(T)}{\lambda^2 - C} + \frac{D(T)}{\lambda^2 - E} \quad (3)$$

the parameters A through E in Eq. (3) were found for each temperature, and in this case, the values of A , B , and D could be fit to quadratic expressions in T with good accuracy. These expressions are provided in Table 2 below. We note that the poles (at $\lambda = 301.83$ nm and 27.53 μm) are close to the poles of 345 nm and 27.5 μm in the Parsons and Coleman Sellmeier equation (Eq. (1)). The difference between the experimental results shown in Fig. 3(a) and the values obtained from Eq. (3) is the fit error Δn (shown in Figs. 3(b)–3(d)).

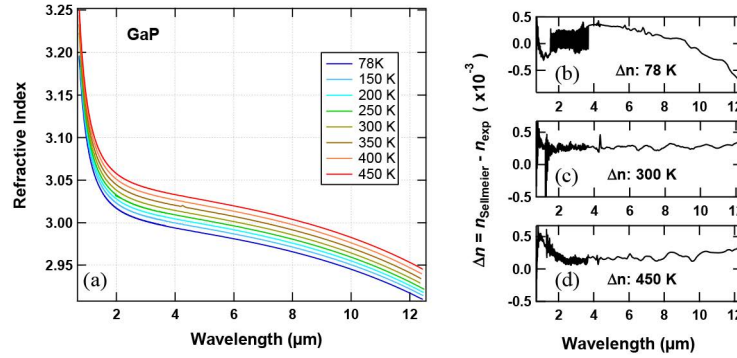


Fig. 3. (a) Temperature and wavelength dependent refractive indices of GaP, in 50 K steps; $n(\lambda, T)$ increases with temperature. (b, c, d) Fit errors between the Sellmeier expression and the experimental refractive index at different temperatures.

Table 2. Temperature-dependent Sellmeier fit coefficients^a

Coef.	n
A	$10.926 + 7.0787 \times 10^{-4} T + 1.8594 \times 10^{-7} T^2$
B	$0.53718 + 5.8035 \times 10^{-5} T + 1.9819 \times 10^{-7} T^2$
C	0.0911014
D	$1504 + 0.25935 T - 0.00023326 T^2$
E	758.048

^aThe temperature T is in Kelvin and wavelength λ is in μm .
The fits are valid for a wavelength range of 0.7 to 12.5 μm ,
for temperatures between 78 and 450 K.

4. Comparison with nonlinear experiments

The temperature dependent Sellmeier equation for GaP, given by Eq. (3) and Table 2, was used to calculate the signal and idler wavelengths generated by frequency conversion of three different pump wavelengths in OP-GaP crystals with three different grating spacings. Figure 4 shows the predicted values (solid lines) and the experimentally measured values (symbols). The predicted and measured values were in good correspondence with one another, indicating that the Sellmeier equation is reliable. (This comparison to experiment validates the wavelength- and temperature-dependence of Eq. (3), although it should be pointed out that adding a wavelength-independent offset in the refractive index will yield the same phase matching curves.) The deviation from experiment was small in all cases, but it was most pronounced in the case of the 1.559 μm pump. The predicted wavelengths are indeed sensitive to refractive index, the pump wavelength, and the grating spacing, and a <0.3% error in any combination of these would account for the slight discrepancy.

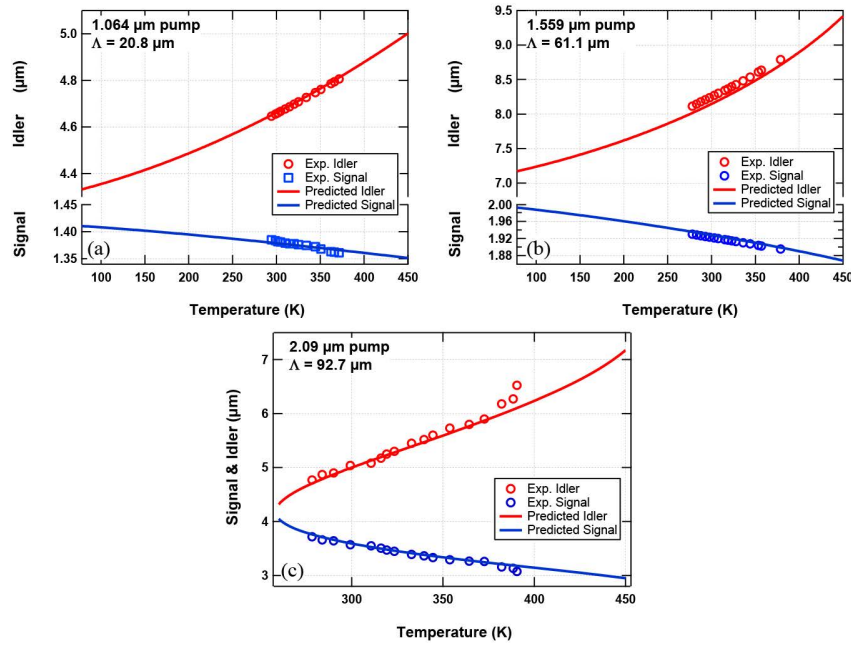


Fig. 4. Comparison between predicted signal and idler wavelengths (solid line) and the experimentally measured values (symbols) for (a) 1.064 μm pump OPO, with $\Lambda = 20.8 \mu\text{m}$, (b) 1.559 μm pump OPO, with $\Lambda = 61.1 \mu\text{m}$, and (c) 2.09 μm pump OPO, with $\Lambda = 92.7 \mu\text{m}$.

5. Summary

The temperature dependent Sellmeier equation for GaP was obtained over the wavelength range 0.7 to 12.5 μm , and over the temperature range 78 K to 450 K for the first time, significantly extending the room temperature results of Parsons and Coleman [2]. A good match was obtained between the temperature dependent signal and idler wavelengths predicted by the equation and the experimentally measured values for three pump wavelengths and grating spacings.

Acknowledgements

The authors thank Dr. Paulina Kuo, NIST, for bringing Ref. [6] and related work to their attention.